# Surface-induced photorefractivity in twistable nematics: toward the all-optical control of gain

P. Pagliusi\*, C. Provenzano and G. Cipparrone

Laboratorio Regionale LICRYL INFM-CNR and CEMIF.CAL – Department of Physics, Università della Calabria, 87036 Rende (CS), Italy \*Corresponding author: pagliusi@fis.unical.it

Abstract: We report the first two-beam coupling investigation of the surface-induced photorefractive effect (SIPRE) in optically twistable nematic liquid crystal cell. The unique space-charge field of SIPRE is exploited to achieve optical tuning of the photorefractive gain. A reconfigurable photoaligning substrate is used to adjust the twist angle, which is proved to be a control parameter for the photorefractive gain. The amplitude of the optical modulation increases gradually with the twist. Its phase shift changes from  $0^{\circ}$  to  $90^{\circ}$  with the polarization state of the two interfering beams. These results pave the way to the all-optical control of the photorefractive gain.

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OCIS codes: (050.1950) Diffraction gratings; (160.5320) Photorefractive materials; (230.3720) Liquid-crystal devices; (240.6670) Surface photochemistry.

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#98498 - \$15.00 USD Received 15 Jul 2008; revised 29 Aug 2008; accepted 31 Aug 2008; published 29 Sep 2008 13 October 2008 / Vol. 16, No. 21 / OPTICS EXPRESS 16343 (C) 2008 OSA

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### 1. Introduction

Organic materials with photo-tunable properties are attractive for optical information technology, because of their structural and chemical flexibility, remarkable performance, and low cost of processing [1]. Among them, photorefractive (PR) organic media and their unique nonlinear optical properties have been extensively investigated over the last years, driven by feasible applications in dynamic holography, phase conjugation, image and optical data processing and amplification [2-5]. Photorefractivity takes place in electrooptic and photoconductive materials where an inhomogeneous light intensity, like the interference pattern established by two coherent beams, results in a spatial modulation of the refractive index [6]. This occurs when photogenerated charge carriers redistribute over macroscopic distances, producing a internal space-charge electric field, which in turn modulates the refractive index. A distinctive feature of the PR effect is the spatial phase-shift  $\varphi_{a}$  between the refractive index grating and the light intensity pattern, which stems from the charge transport mechanisms in the material (i.e.,  $\varphi_p \sim 90^\circ$  for a pure diffusive charge transport). This non-local photo-response can lead to asymmetric energy transfer (AET) in the two-beam coupling (TBC) configuration, i.e. an exchange of energy between the two optical beams which interfere in the PR medium. The TBC process is at the basis of many proposed applications of PR media, including self-phase conjugation, optical limiting, all-optical image and signal amplification [7,8]; therefore, continuous efforts have been made to enhance and conveniently adjust the AET. The control of the AET has been investigated in the field of classic inorganic photorefractivity, where it has been accomplished adjusting either the refractive index amplitude or the spatial phase-shift  $\varphi_p$ . Different methods have been explored, such as

applying an external ac voltage [9,10], moving the holographic grating [11], varying the intensity of the beams, adopting a double two-wave mixing scheme [12,13].

Within the field of organic PR materials, PR nematic liquid crystals (NLCs) have been widely studied, mainly because they achieve higher TBC gain for electric field values that are orders of magnitude lower compared to PR polymers and organic crystals [14, 15]. The NLCs are fluids with large dielectric, conductive and optical anisotropy, which can be easily aligned in different director configurations (i.e., homogeneous, homeotropic, hybrid, twisted, etc.) thanks to their sensitivity to surface anchoring conditions [16]. Indeed, their high spontaneous birefringence and high dielectric anisotropy lead to a large refractive index modulation through the electric field-induced reorientation of the NLC director (orientational enhancement). Nevertheless, liquid crystalline materials are usually not intrinsically photoconductive, therefore the space-charge field set-up requires either doping them with a photo-charge generating and transporting agents (bulk photorefractivity) [17-20], or confining the NLC layer between photoconducting or photorefractive materials (surface photorefractivity) [21-29]. In bulk photorefractivity, charge carriers are generated and redistribute directly in the LC volume. On the contrary, in surface photorefractivity, the space-charge electric field originates from a redistribution of charges in the confining/aligning substrates (i.e., PR crystal slabs, photoconductive polymers, etc.) or at the interface between the NLC and the confining layers, even if they are not intrinsically photoconductive [27-29].

The surface PR effects in NLCs have recently gained increasing relevance because of a wider range of practical phenomena [21-29], which yield superior performances [23] and a greater flexibility in the device design, through the proper choice of director configuration and materials (NLC, dopants, substrates, etc.). Large PR gain even in the Bragg regime has been demonstrated using PR inorganic crystal slabs to confine a planar aligned NLC layer doped with ferroelectric nanoparticles [25]. A PR gain of constant sign under ac applied voltage, which is desirable to reduce deterioration of NLC cells, has been achieved in an asymmetric hybrid NLC cell, where a single photoconducting film is present, adjacent to the planar aligning substrate [27]. The "on-off" switch of the AET has been recently demonstrated through the proper exploitation of the space-charge electric field of the surface induced PR effect (SIPRE) in a planar NLC cell, by varying both the beams polarization state and the relative orientation between the cell axis and the grating vector [28]. Recently, the SIPRE has been exploited in a non-holographic configuration in NLC twist cell, where the optical control of the Friedericksz transition has been studied for optical limiting applications [30].

The aim of the present work is to provide a viable route to control the PR gain, independently from the intensity and the phase of the two interfering beams. Here, we report the first observation and characterization of holographic surface photorefractivity in TNs and the first example of all-optical control of the steady-state AET via the SIPRE in NLCs. Our approach is based on the peculiar space-charge field produced by the SIPRE and on the anisotropic orientational response of the NLC to the electric field. In fact, regardless of the exact nature of the photoelectric processes, which are responsible of the surface-charge distribution in the SIPRE, the resulting space-charge electric field in the NLC layer is characterized by two orthogonal modulated components: a transverse component (along the grating vector) which is 90° phase-shifted with respect to the interference pattern, and an inphase longitudinal component (perpendicular to the grating vector, along the cell normal) [31]. To overcome the limitations imposed by the planar NLC configuration with regard to the control of the PR gain [28], we move the nematic director out of the in-plane arrangement and choose the twisted-nematic (TN) configuration. In particular, we adopt an asymmetric cell configuration, where one substrate is a rubbed polymer film, which provides a stable homogeneous alignment, the other is a photoaligning layer, which allows to adjust the twist angle  $\psi$  optically. When irradiated with proper linearly polarized light, in fact, the photoaligning substrate provides a reconfigurable homogenous alignment of the NLC [32, 33]. For this reason, we indicate the system under investigation as optically twistable NLC cell. Indeed, we prove that the twist angle  $\psi$  and the polarization state of the interfering beams

are independent control parameters for the PR gain coefficient and, therefore, allow for fine tuning of the AET.

#### 2. Experimental results and discussion

In our sample, the NLC layer is sandwiched between two different polymer-coated indium tin oxide (ITO) glass slides, as shown in Fig. 1. The central 25µm-thick layer consists of the eutectic NLC E7 (BL001 from Merck KGaA, Darmstadt), which is nematic at room temperature (Cr -20°C N 61°C I). It is a mixture of four cvano-substituted polyphenyls: 4cyano-4'-n-pentyl-biphenyl (5CB, ~51%), 4-cyano-4'-n-heptyl-biphenyl (7CB, ~25%), 4cyano-4'-n-oxyoctyl-biphenyl (80CB, ~16%), and 4-cyano-4"-n-pentyl-p-terphenyl (5CT,  $\sim$ 8%). In order to provide a reference planar anchoring for the NLC, a 20nm-thick layer of polyvinyl alcohol (PVA, 87% hydrolyzed, M.W. 31000-50000, from Aldrich) is spin-coated from its water solution (1% by weight) on one of the two ITO-substrates. The substrate is baked for 60 minutes at 120°C to eliminate the solvent, slowly cooled to room temperature and then rubbed uniaxially using a velvet-wrapped roller to induce planar alignment. On the other substrate, a 20 nm-thick polarization sensitive photoaligning command layer is spincoated from a water solution of PVA (1% by weight) and a dichroic azobenzene dye (0.5%), by weight), and baked for 60 minutes at 120°C. The azo-dye, whose chemical formula and absorption spectra are reported in Ref. 34, belongs to the class of sulphuric bis-azobenzene dyes which are drawing interest as photoaligning agent for low-molecular-weight LCs, polymeric and polymerizable LCs, because of their photosensitivity and high thermal, photochemical and electrochemical stability [35]. A remarkable property of this class of azodye is the pure orientation of the molecular absorption oscillators perpendicular to the polarization of the activating light, without photochemical transformations like *cis-trans* isomerization [36]. The cell is assembled by placing 25µm-thick Mylar spacers between the two substrates and the E7 is infiltrated by capillary action in the isotropic phase ( $T = 68^{\circ}$ C) and then slowly cooled to room temperature. Due to the negligible azimuthal anchoring energy of the unexposed photoaligning substrate, an homogeneous planar cell is obtained ( $\psi =$  $0^{\circ}$ ), where the LC director is oriented along the anchoring direction of the rubbed PVA substrate (y-axis in Fig. 1).

The TN cell configuration is obtained inducing a preferential alignment direction (*easy axis*) for the LC molecules adsorbed on the photoaligning substrate, which deviates from the y-axis. It can be achieved exposing the cell from the photoaligning side (see Fig. 1) to a linearly polarized beam of proper wavelength for a dose of 1-10 J/cm<sup>2</sup>. In the experiment, the control over the angle of twist  $\psi$  is performed with the Ar<sup>+</sup>-ion laser ( $\lambda = 457.8$ nm) beam c of intensity  $I_c \cong 50$ mW/cm<sup>2</sup>, according to the following procedure. The linear polarization of the beam c is first oriented along the x-axis to start the *easy axis* along the y-axis. Then, with the help of a half-wave plate, the polarization is gradually rotated clockwise (CW) or anticlockwise (ACW) of the appropriate angle, and finally kept for few minutes to strengthen the NLC anchoring. According to this procedure, it is possible to obtain single-domain TN cell (i.e., uniform handedness) even for  $|\psi| \ge 90^\circ$ . The resulting twist angle value  $\psi$  (positive or negative) is easily estimated from the adiabatic rotation (CW or ACW) of the polarization plane of the counter-propagating, y-polarized He-Ne ( $\lambda = 632.8$ nm) probe beam d (see Fig. 1).



Fig. 1. Experimental layout showing the NLC cell, the interfering beams (*a* and *b*), the photoaligning (*c*) and the probe (*d*) linearly polarized beams (black dotted arrows indicate the polarization direction). The NLC cell is made of two ITO substrates: the right one is the photoelectrically activable substrate, coated with a PVA film (yellow) and uniaxially rubbed to induce planar alignment along the y-axis; the left one is the photoaligning layer (orange), coated with the azo-dye-doped PVA.  $\psi$  indicates the twist angle, measured from the negative y-axis.  $V_{DC}$  is the polarizing dc voltage applied to the ITO electrodes. The red and blue spheres represent the anions and cations accumulated at the anodic and cathodic interface, respectively.  $E_x$  and  $E_z$  are the transverse and longitudinal space-charge electric field components in the NLC layer, respectively.

As usual in SIPRE experiments, a dc voltage ( $V_{dc}$ = 4.0V) is applied to the ITO electrodes before the sample irradiation, to collect charge carriers (ions due to dissociated LC molecules and/or residuals of chemical synthesis) of opposite sign at the polymer-LC interfaces. Two parallel polarized Ar<sup>+</sup>-ion laser beams ( $\lambda = 457.8$ nm) of equal intensity ( $I_0 \cong 0.5$ mW/cm<sup>2</sup>) interfere on the sample in a non-slanted geometry, i.e. the cell normal is parallel to the beams bisector, producing the intensity modulation  $I(x) \propto 1 - cos(q \cdot r) = 1 - cos(qx)$ , where  $q = qe_x$  is the grating vector and  $\Lambda = 2\pi/q \cong 28\mu m$  is the spatial periodicity ( $Q = 2\pi\lambda d/\Lambda^2 n_0 \ll 1$ , thin grating regime). The beams impinge on the cell from the rubbed PVA side (right side in Fig. 1). Photoinduced modulation of the surface charge density mainly occurs at the anodic PVA-E7 interface, yielding to a space-charge electric field in the NLC layer having both the  $E_x$  and  $E_z$  components spatially modulated along q, with full periodicity, and phase equal to 90° and 0° with respect to I(x), respectively [31].

The optical modulation, resulting from the space-charge-field-induced director distortion, and the corresponding AET are investigated with TBC measurements by the gratingtranslation technique [2]. The technique allows to determine the effective amplitudes  $(\Delta n, \Delta K)$ and spatial phases ( $\varphi_p$ ,  $\varphi_a$ ) of both the refractive index and the absorption coefficient modulations, i.e.  $n(x) = n_0 + \Delta n \cos(qx \cdot \varphi_p)$  and  $K(x) = K_0 + \Delta K \cos(qx \cdot \varphi_a)$ . After the grating recording, the sample is rapidly translated along the grating vector and the transmitted intensity of the two self-diffracting beams ( $I_1$  and  $I_2$ ) is measured versus the grating shift  $\delta$ . The shifting speed is 500µm/s, high enough to avoid grating perturbation during the measurement. In the low diffraction efficiency regime ( $\leq 10\%$ ), the sum  $I_+(\delta) \equiv I_1 + I_2 \cong I_0$  $exp(-K_0d) [2-4A\cos(\varphi_a + q\delta)]$  and the difference  $I_{-}(\delta) \equiv I_1 - I_2 \cong I_0 exp(-K_0d) [-4P\sin(\varphi_p + q\delta)]$ of the intensities are both harmonic function of  $\delta$ , whose amplitudes are proportional to  $A \cong$  $\Delta Kd/4$  and  $P \cong \pi \Delta n d/\lambda$  respectively, where d is the sample thickness. The NLC being transparent in the whole visible range, no significant modulation of the absorption coefficient occurs ( $A \propto \Delta K \cong 0$ ), therefore  $I_{+}(\delta) \cong 2I_0 \exp(-K_0 d)$  is nearly independent of  $\delta$  [28]. From the  $I_{+}(\delta)$  curve,  $I_{0} exp(-K_{0}d) = I_{+}(\delta)/2$  has been easily calculated. Then,  $\Delta n$ ,  $\varphi_{p}$  and the exponential gain coefficient  $\Gamma = (4\pi/\lambda) \Delta n \sin \varphi_p$  have been evaluated from the  $I_{\cdot}(\delta)$  curve, normalized versus  $I_0 exp(-K_0 d)$ .

In Fig. 2 we report the normalized  $I_{\cdot}(\delta)$  curves, resulting from the TBC measurements of *p*-polarized beams, for different values of the twist angle  $\psi$  in the range 0-90°. For the sake of figure clarity, only the  $I_{+}(\delta)$  for  $\psi = 90^{\circ}$  has been reported in the inset, being similar to the  $I_{+}(\delta)$  curves for all the other  $\psi$  values. The  $I_{+}(\delta)$  is constant versus  $\delta$ , confirming the development of pure phase gratings in the NLC cell.



Fig. 2. Normalized  $I_{.}(\delta)$  curves obtained from TBC measurements on the TN cell, for *p*-polarized beam and different values of the twist angle  $\psi$ . In the inset, the  $I_{+}(\delta)$  curve (red dots) for  $\psi = 90^{\circ}$  is reported.

The modulation amplitude (4P) of the normalized  $I_{\cdot}(\delta)$  curves is proportional to the effective refractive index modulation  $\Delta n$  experienced by the interfering beams. For  $\psi = 0^{\circ}$  (homogenous cell), the p-polarized beams do not experience diffraction, i.e.  $\Delta n \cong 0$ , which is in agreement with previous observations [28]. When  $\psi$  increases (TN cell), the effective refractive index modulation  $\Delta n$  grows up monotonically to  $5 \times 10^{-4}$  (for  $\psi = 90^{\circ}$ ), while the phase shift  $\varphi_p$  remains equal to 90° within the experimental error, in the whole explored range (see Table 1).

Ψ	$\Delta n$	$arphi_p$	$\Gamma(cm^{-1})$
0°	0	undefined	0
30°	$(1.1 \pm 0.2) \times 10^{-4}$	(90 ± 20)°	30
60°	$(3.2 \pm 0.5) \times 10^{-4}$	$(70 \pm 20)^{\circ}$	80
90°	$(5.0 \pm 0.5) \times 10^{-4}$	$(90 \pm 20)^{\circ}$	140

Table 1. Effective refractive index modulation parameters (amplitude  $\Delta n$  and phase  $\varphi_p$ ) and PR gain coefficient  $\Gamma$  for different values of the twist angle  $\psi$ 

Accordingly, the PR gain coefficient  $\Gamma$  of the *p*-polarized beams, which is zero for the homogenous cell ( $\psi = 0^{\circ}$ ), increases with  $\psi$  up to 140cm<sup>-1</sup>, keeping constant both the beams intensity ( $I_0 \cong 0.5 \text{mW/cm}^2$ ) and the dc voltage ( $V_{dc} = 4.0 \text{V}$ ) applied to the cell. As a result, the twist angle  $\psi$  plays the role of control parameter for the  $\Gamma$  and, then, for the AET.

Control of the twist angle  $\psi$  is not the unique approach to optically tune the PR gain in TN cells exploiting the SIPRE. Indeed, we might keep the TN cell configuration at constant  $\psi$  and vary the polarization state of the interfering beams. Optical modulation in birefringent medium is generally strongly anisotropic and the SIPRE in TN cells does not represent an exception, even though we do not expect the extreme optical anisotropic (on-off) behaviour of the homogenous cells [31]. In Fig. 3 we report the normalized  $L(\delta)$  curves  $(L(\delta)/I_0 exp(-K_0d))$  resulting from the TBC analyses on the  $\psi = 90^{\circ}$  TN cell, for different linear polarization states of the interfering beams, namely *p*- and *s*-polarized beams, keeping constant all the other parameters  $(I_0, V_{dc}, q)$ . In both the cases, the interfering beams experience diffraction because of an effective refractive index modulation, whose amplitude  $\Delta n$  is comparable for the two polarization states (~5.0×10<sup>-4</sup>). On the other hand, it is remarkable that moving from *p*- to *s*-polarized beams, the phase shift changes from  $\varphi_p \sim 90^{\circ}$  to  $0^{\circ}$  and, accordingly, the PR gain coefficient changes from its maximum value  $\Gamma \cong 140 \text{ cm}^{-1}$  to  $0\text{ cm}^{-1}$ .



Fig. 3. Normalized  $I_{.}(\delta)$  curves obtained from TBC measurements on the  $\psi = 90^{\circ}$  TN cell, for *p*-polarized and *s*-polarized interfering beams. The modulation amplitude, shown by the red bar, is equal to  $4\pi\Delta nd/\lambda$ .

The experimental results reported here demonstrate that, exploiting the TN configuration and the SIPRE, two different strategies have been identified which allow for optical control of the PR gain. First, it has been accomplished via fine tuning of the effective grating amplitude  $\Delta n$ , as a result of the optical manipulation of the twist angle  $\psi$ . Second, the anisotropy of the modulated director distribution in the TN cell has been exploited to switch the phase  $\varphi_p$  of the effective grating between 0° and 90°, by means of the polarization state of the interfering waves.

The occurrence of AET between the recording beams is a clear indication of the PR origin of the optical modulation, produced in the NLC cell via periodic director distribution. In particular, the latter stems from the SIPRE space-charge electric field, characterized by spatially modulated  $E_x$  and  $E_z$  components, having phases equal to 90° and 0° with respect to I(x), respectively [28]. Beside the twist angle value, the space-charge electric field and the director configuration depend only on the optical intensity modulation I(x) on the photoelectrically activable PVA-E7 interface. Given the small angle ( $\theta \sim 1^\circ$ ) between the interfering beams, I(x) is independent of the (s- or p-) polarization state of the interfering beams. However, because of the significant NLC birefringence, the effective optical modulation and, therefore, the coupling behaviour are strongly affected by the polarization state of beams, as shown in Fig. 3.

A thorough understanding of the experimental findings reported here requires to determine the absolute director distribution in the TN configuration, through the minimization of the nematic free-energy, taking into account the elastic, dielectric and surface (anchoring) contributions. This is beyond the scope of the present paper and will be reported elsewhere. Nevertheless, we can give an insight into them, starting from the analysis of the corresponding SIPRE experiments performed in homogeneous NLC cells [28]. Significant AET occurs if the optical axis of the planar cell is parallel to the grating vector, while no energy coupling takes place rotating the optical axis perpendicular to it. Indeed, the TBC analysis demonstrated a 90° phase-shifted refractive index modulation ( $\varphi_p \sim 90^\circ$ ) in the first geometry, and an in-phase grating  $(\varphi_p \sim 0^\circ)$  for the second geometry. Moreover, the resulting optical modulations are in both cases strongly anisotropic, signifying that the nematic director reorientation is always confined within the plane normal to the cell surfaces and parallel to the rubbing direction [31]. This is particularly interesting for the planar configuration in which the optical axis is perpendicular to the grating vector, which is equivalent to the one depicted in Fig. 1 for  $\psi = 0^{\circ}$ . In this case, we found the director to be confined in the yz-plane, then the sole longitudinal space-charge electric field component  $E_z$  is responsible for the optical grating. The transverse component, which would induce a 90° phase-shifted twist director deformation, actually does not influence the nematic director. Accordingly, p-polarized beams do not experience any effective optical modulation, as confirmed here by the TBC curve  $(\psi = 0^{\circ})$  in Fig. 2. Moving the NLC cell from a homogeneous to a TN configuration, as illustrated in Fig. 1, p-polarized beams pass through a refractive index modulation of increasing amplitude versus  $\psi$ , but constant phase shift  $\varphi_p \sim 90^\circ$  (see Table 1). This result suggests that twisting the director configuration out of the yz-plane, a further spatially modulated twist is allowed. It is driven by the transverse electric field component  $E_x$  and produces the  $90^{\circ}$  phase-shifted grating experienced by the *p*-polarized beams.

It is worth noting that, even in the TN configuration, the *s*-polarized beams still feel an inphase ( $\varphi_p \sim 0^\circ$ ) effective refractive index modulation (see Fig. 3), in the same way as for the homogenous configuration ( $\psi = 0^\circ$  in Fig. 1) [28].

#### 3. Summary

In the present paper we demonstrate the possibility to obtain a pure optical control of the PR gain, exploiting the SIPRE in twistable nematic configuration for the first time. An asymmetric cell configuration has been adopted, where one substrate is a photoaligning layer, which allows for reconfigurable optical adjustment of the twist angle  $\psi$ . Two strategies have been identified according to which the twist angle and the polarization state of the interfering beams are independent control parameters for the PR gain coefficient. First, the PR gain between two interfering *p*-polarized beams progressively increases with  $\psi$ , starting from zero when the cell is planar ( $\psi = 0^{\circ}$ ). In fact, the effective optical modulation amplitude  $\Delta n$  grows from zero to its maximum value, while the phase-shift remains almost constant ( $\varphi_p \sim 90^{\circ}$ ). Second, the spatial phase-shift  $\varphi_p$  of the effective optical modulation, and therefore the PR gain coefficient, depends on the polarization state of the interfering beams. In particular, we

show that for  $\psi = 90^\circ$ , the spatial phase shift  $\varphi_p$  changes from 90° to 0° switching from *p*- to *s*-polarized beams.

## Acknowledgments

The authors thank Prof. R. Bartolino and Prof. L. M. Blinov for fruitful discussions and Dr. Alfredo Pane for cell preparation in clean-room facility.